

#### Final Report on Program:

# Development of Materials and Laser Devices in the Fiber Configuration

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#### **Brief Summary**

#### **Program Goals:**

 Development of both new and optimized materials using the laser heated pedestal growth technique

#### **Applications:**

- New laser materials
- Diode pumped fiber laser devices

#### Accomplishments:

- Growth of both fluoride and oxide single crystal fibers with a range of dopants and concentrations
- Investigation of Cr<sup>4+</sup> lasers and materials
- Three dimensional simulations of LHPG
- Dynamics of upconversion and avalanche processes
- Ultranarrow spectral holeburning in LHPG fibers
- Phonon Transport in LHPG fibers
- Diode pumped laser action in LHPG fibers

#### **Technical Summary**

In this section we provide a brief description of the major thrusts, goals and achievements of our program. More detailed information is available in the publications which resulted from this program. A complete list of the publications, proceedings and papers presented as a result of this work are given in Appendix II.

Growth of both fluoride and oxide single crystal fibers with a range of dopants and concentrations:

Over this grant period we have been continuously developing and optimizing the Laser Heated Pedestal Growth (LHPG) technique. In order to facilitate the growth of fluoride single crystal fibers we have developed a hydrofluorinator for the preparation of precursor materials and adapted our original LHPG station to operate under vacuum. We have also designed and contracted a second station which includes an improved control system, and improved stability and monitoring. Using these facilities we have grown over a hundred materials a complete listing of which is provided in Appendix I

#### Investigation of Cr<sup>4+</sup> lasers and materials:

We have performed detailed static and dynamic spectroscopic investigations of Cr<sup>4+</sup> doped materials. These investigations include a determination of the polarization dependence of the saturation in this material under different crystal orientations. These results provide an explanation as to why this material is more effectively longitudinally pumped by light polarized along the 100 axis. We have also performed extensive laser characterization of this system under both pulsed and continuous wave pumping.

#### Three dimensional simulations of LHPG:

We have performed molecular dynamics simulation of the laser heated pedestal growth techniques. All particles in the simulation interact according to a Lennard-Jones type potential. This work demonstrates that it is possible to simulate this process on a microscopic scale. Behavior observed in the simulations includes: fracturing, bubble formation and the spatial evolution of tagged particles in the source rod. The flow patterns in the liquid which during the pulling process were also determined. In addition to the above results, the structure factor as a function of position in phase space was calculated.

#### Dynamics of upconversion and avalanche processes:

We have investigated upconversion processes in both YAG:Pr<sup>3+</sup> and Y<sub>2</sub>O<sub>3</sub>:Tm<sup>3+</sup> single crystal fibers. In both YAG:Pr<sup>3+</sup> ultraviolet upconversion emission was In the latter system blue upconversion was observed to occur by the avalanche mechanism. We have performed additional studies in the YLF:Nd<sup>3+</sup> system, where both green and violet upconversion was observed under infrared pumping. We were able to establish that Nd pairs were instrumental in the production of both upconversion processes.

#### Optical dephasing and ultranarrow spectral holeburning in LHPG fibers:

Optical dephasing of rare earth ions in crystalline solids has been studied both in the time and frequency domain to examine. These studies explore dynamical processes which occur both inherently in the ideal crystal and those due to defects or disorder. These studies have enabled us to understand the optical dephasing of paramagnetic ions due to superhyperfine interactions with the nuclear moments of the host nuclei. They have also shown that time domain spectroscopies can be extremely sensitive probes of the presence

of defects which contribute to the system's dynamics. Effects of both UV-induced defects and of defects introduced in the growth process using the Laser-Heated Pedestal Growth (LHPG) have been demonstrated. Finally, we have observed the narrowest spectral holes in a solid using an ultranarrow (1kHz) bandwidth laser. This laser has made it possible to observed time-dependent optical holeburning, allowing us to observe very weak spectral diffusion which occurs due to defects in the LHPG samples.

#### Phonon Transport in LHPG fibers:

We have investigated the transport of nonequilibrium phonons in both YAG and ruby single crystal fibers. We have in the YAG system we determined the modes of propagation and examined the cross over between three dimensional and one dimensional transport down the fiber length. In ruby the fiber geometry enabled us to determine the contribution of surface scattering to the phonon decay rate. In addition when immersed in superfluid helium, the single crystal fiber was shown to behave as a wave vector filter.

#### Diode pumped laser action in LHPG fibers:

We have observed diode pumped laser action in Nd:YAG fibers grown by the University of Georgia LHPG facility. Fibers with a Nd concentrations of 2% and 5% were cut and polished to 3µm, annealed at 1100°C for 20 hours and anti reflection coated for 1.064 µm. The 5% fiber exhibited a lasing threshold at 80 mW with a slope efficiency of 18% while the 2% sample exhibited a lasing threshold at 65 mW with a slope efficiency of 25%. A commercial crystal with 1% Nd<sup>3+</sup> the same pumping configuration sample exhibited a lasing threshold at 40 mW with a slope efficiency of 40%.

## Appendix I

Materials Grown with the University of Georgia LHPG system

## Materials Grown with UGA LHPG System

Al<sub>2</sub>O<sub>3</sub>

doped with:

Cr3+,

Ti3+,

Cr3+ and Ti3+,

Mg<sup>2+</sup>,

Si<sup>4+</sup>,

Cr4+ and Si4+,

Ti<sup>2+</sup> and Si<sup>4+</sup>,

Mg<sup>2+</sup> and Cr<sup>4+</sup>,

 $Mg^{2+}$  and  $Mn^{4+}$ ,

Co<sup>2+</sup> and Si<sup>4+</sup>

BaTiO<sub>3</sub>

doped with:

Eu3+

BaYFa

doped with:

Er3+

CaF<sub>2</sub>

undoped

doped with:

Pb<sup>2+</sup>,

CaWO<sub>4</sub>

doped with:

doped with:

Ti<sup>2+</sup>

CsB<sub>3</sub>O<sub>5</sub>

undoped

Pr3+

DyF<sub>3</sub>

GdEuO<sub>3</sub>

doped with:

Nd3+

**GGG** 

doped with:

Cr34

$Gd_2O_3$	doped with:	Nd <sup>3+</sup>
GdScO <sub>3</sub>	doped with:	Nd <sup>3+</sup>
GGAG	doped with:	Ca <sup>2+</sup> and Cr <sup>4+</sup>
GSAG	doped with:	Ca <sup>2+</sup> and Cr <sup>4+</sup> , Mg <sup>2+</sup> and Cr <sup>4+</sup>
LaAlO <sub>3</sub>	doped with:	Cr <sup>3+</sup> and Eu <sup>3+</sup>
LaF <sub>3</sub>		
LaGaGeO <sub>7</sub>	doped with:	Nd <sup>3+</sup>
La <sub>3</sub> Ga <sub>5</sub> SiO <sub>14</sub>		: :
LIAI508	doped with:	Ni <sup>2+</sup> , Co <sup>2+</sup>
LiCaAIF <sub>6</sub>	doped with:	Cr <sup>3+</sup>
LiF		
LiGa <sub>5</sub> O <sub>8</sub>	doped with:	Co <sup>2+</sup> , Ni <sup>2+</sup> , Co <sup>2+</sup> and Mg <sup>2+</sup>
Linbo <sub>3</sub>		
LIYF4	doped with:	Er <sup>3+</sup>
MgAl <sub>2</sub> O <sub>4</sub>	doped with:	Cr <sup>3+</sup> ,

Ti<sup>3+</sup>

### MgCaSiO<sub>4</sub>

 $\mathrm{Mg}_{1.5}\mathrm{Mn}_{0.5}\mathrm{SiO}_{4}$ 

doped with:

Cr<sup>4+</sup>

Mg<sub>2</sub>SiO<sub>4</sub>

doped with:

Cr3+ and Cr4+

Mn<sub>2</sub>SiO<sub>4</sub>

NaLa(WO<sub>4</sub>)<sub>2</sub>

doped with:

Er3+,

Eu<sup>3+</sup>,

Nd3+,

Er3+ and Yb3+

NaY(WO<sub>4</sub>)<sub>2</sub>

doped with:

Eu<sup>3+</sup>

NdF<sub>3</sub>

PbMoO<sub>4</sub>

RbMnF<sub>3</sub>

Sc<sub>2</sub>O<sub>3</sub>

doped with:

Er3+,

Nd<sup>3+</sup>,

Ti3+

SrAl<sub>2</sub>O<sub>4</sub>

doped with:

Cr<sup>4+</sup>

SrTiO<sub>3</sub>

doped with:

Eu3+,

Nd<sup>3+</sup>,

Cr3+

**YAG** 

doped with:

Ca<sup>2+</sup>,

Ce<sup>3+</sup>,

Cr3+,

Er3+,

Nd3+,

Pr3+,

Ti3+,

Tm<sup>3+</sup>,

v<sup>3+</sup>,

Ca<sup>2+</sup> and Mn<sup>4+</sup>,

 $Co^{2+}$  and  $Si^{4+}$ ,

 ${\rm Mg^{2+}}$  and  ${\rm Mn^{4+}}$ ,

Ti<sup>3+</sup> and Nd<sup>3+</sup>, Ti<sup>2+</sup> and Si<sup>4+</sup>,

Tm<sup>3+</sup> and Ce<sup>3+</sup>

YALO<sub>3</sub>

doped with:

Er3+,

Er3+ and Eu3+

**YGAG** 

doped with:

Ca<sup>2+</sup> and Cr<sup>4+</sup>

YGG

doped with:

Mg<sup>2+</sup> and Cr<sup>4+</sup>

YLF

Y<sub>2</sub>O<sub>3</sub>

doped with:

Ce<sup>3+</sup>,

Dy<sup>3+</sup>,

Er3+,

Eu3+,

Ho<sup>3+</sup>,

Nd3+,

Pr3+,

Tb3+,

Tm<sup>3+</sup>,

Dy3+ and Tb3+,

Pr3+ and Yb3+,

Tm<sup>3+</sup> and Yb<sup>3+</sup>

YScO<sub>3</sub>

undoped

doped with:

Er3+,

Eu3+,

Nd3+

Y<sub>1.96</sub>Sc<sub>0.04</sub>O<sub>3</sub>

doped with:

Eu3+

Y<sub>1.60</sub>Sc<sub>0.40</sub>O<sub>3</sub>

doped with:

Eu<sup>3+</sup>

Y2SIO5

doped with:

Eu3+

YSAG

doped with:

Ca<sup>2+</sup> and Cr<sup>4+</sup>

**YSGG** 

doped with:

Mg<sup>2+</sup> and Cr<sup>4+</sup>

ZrSiO<sub>4</sub>

doped with:

Cr<sup>4+</sup>

Appendix II

**Publications** 

#### Publications: Refereed Journals

- "Saturation of the 1.064mm Absorption in Cr<sub>1</sub>Ca:Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub> Crystals," H. Eilers, K.R. Hoffman, W.M. Dennis, S.M. Jacobsen and W.M. Yen, Appl. Phys. Lett. <u>61</u>, 2958-2960 (1992).
- "Performance of a Cr:YAG Laser," H. Eilers, W.M. Dennis, W.M. Yen, S. Kück, K. Petermann, G. Huber and W. Jia, IEEE J. Quantum Electron. 29, 2508-2512 (1993).
- "Spectroscopic Properties of Cr<sup>4+</sup>:Lu<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>," H. Eilers, U. Hömmerich, S.M. Jacobsen, W.M. Yen and M. Kokta, Opt. Lett. <u>18</u>, 1928-1930 (1993).
- "The Near Infrared Emission of Cr:Mn<sub>2</sub>SiO<sub>4</sub> and Cr:MgCaSiO<sub>4</sub>," H. Eilers, U. Hömmerich, S.M. Jacobsen and W.M. Yen, Chem. Phys. Lett. <u>212</u>, 109-112 (1993).
- "Near Infrared Luminescence Properties of the Laser Material Cr:Y<sub>2</sub>SiO<sub>5</sub>," U. Hömmerich, H. Eilers, S.M. Jacobsen and W.M. Yen, J. Lumin. <u>55</u>, 293-297 (1993).
- "Observation of Avalanche-Like Behaviour in Tm<sup>3+</sup>:Y<sub>2</sub>O<sub>5</sub>," J.M. Dyson, S.M. Jaffe, H. Eilers, M.L. Jones, W.M. Dennis and W.M. Yen, J. Lumin. (in press).
- "Spectroscopy and Dynamics of Cr<sup>4+</sup>:Y<sub>3</sub>Al<sub>5</sub>O<sub>12</sub>," H. Eilers, U. Hömmerich, S.M. Jacobsen and W.M. Yen, Phys. Rev. B (to be published).
- "Infrared to Violet Up-Conversion in YLiF<sub>4</sub>:Nd<sup>3+</sup>," A.M. Novo-Gradac, W.M. Dennis, A.J. Silversmith, S.M. Jacobsen and W.M. Yen, J. Lumin. (in press).
- "Molecular Dynamics Study of the Growth of Optical Fibers," M.J.P. Nijmeijer and D.P. Landau, Computational Materials Science 1, 389-402 (1993).
- "Magnetic Field Dependence of Photon Echo Decays in Ruby," J. Ganem, Y.P. Wang, R.S. Meltzer and W.M. Yen, Phys. Rev. <u>B43</u>, 8599 (1991).
- "Nonexponential Photon-Echo Decays of Paramagnetic Ions in the Superfine Limit," J. Ganem, Y.P. Wang, D. Boye, R.S. Meltzer, W.M. Yen and R.M. Macfarlane, Phys. Rev. Lett. <u>66</u>, 695-698 (1991).
- "Optical Dephasing of Paramagnetic Ions: Er<sup>3+</sup>:YLiF<sub>4</sub>: Experiments and Computer Simulations," R.S. Meltzer, J. Ganem, Y.P. Wang, D. Boye, W.M. Yen, D.P. Landau, R. Wannemacher and R.M. Macfarlane, J. Lumin. <u>53</u>, 80 (1992).
- "Time Resolved Spectral Holeburning in Er<sup>3+</sup>:YLiF<sub>4</sub>: Experiments and Computer Simulations," Y.P. Wang, R.S. Meltzer and R.M. Macfarlane, J. Opt. Soc. Am. <u>B9</u>, 946-949 (1992).

- "Inhomogeneous Broadening by Nuclear Spin Fields: A New Limit for Optical Transitions in Solids," R.M. Macfarlane, A. Cassanno and R.S. Meltzer, Phys. Rev. Lett. <u>69</u>, 542-545 (1992).
- "Modulation of Photon Echo Intensities by Electric Fields: Pseudo-Stark Splittings in Alexandrite and YAlO<sub>3</sub>:Er<sup>3+</sup>," Y.P. Wang and R.S. Meltzer, Phys. Rev. <u>B45</u>, 10119 (1992).
- "Persistent UV-Induced Optical Dephasing in Pr-Doped Yttrium Aluminum Garnet," K.-W. Jang, R.S. Meltzer and J. Ganem, Phys. Rev. <u>B49</u>, 3009 (1994).
- "Sample-Dependent Optical Dephasing in Bulk Crystalline Samples of Y<sub>2</sub>O<sub>3</sub>:Eu<sup>3+</sup>," G.P. Flinn, K.-W. Jang, J. Ganem, M.L. Jones, R.S. Meltzer and R.M. Macfarlane, Phys. Rev. <u>B49</u>, 5821 (1994).
- "Time-Resolved Ultranarrow Optical Hole Burning of a Crystalline Solid: Y<sub>2</sub>O<sub>3</sub>:Eu<sup>3+</sup>," M.J. Sellars, R.S. Meltzer, P.T.H. Fisk and N.B. Manson, J. Opt. Soc. Am. 11, 1 (1994).
- "Persistent Optical Coherence Loss by UV Irradiation in Pr<sup>3+</sup>:YAG," K.-W. Jang, R.S. Meltzer and J. Ganem, J. Lumin. <u>58</u>, 311 (1994).
- "Anomalous Optical Dephasing in Crystalline Y<sub>2</sub>O<sub>3</sub>:Eu<sup>3+</sup>," G.P. Flinn, K.-W. Jang, J. Ganem, M.L. Jones, R.S. Meltzer and R.M. Macfarlane, J. Lumin. <u>58</u>, 374 (1994).
- "Phonon Dynamics in YAG:Pr<sup>3+</sup>," Xiao-jun Wang, W.M. Dennis and W.M. Yen, J. Lumin. <u>53</u>, 44 (1992).

#### Publications: Proceeding Articles

"Dynamics of Nonequilibrium THz Phonons in Single Crystal Ruby Fibers," S.A. Basun, A.A. Kaplyanskii, S.P. Feofilov and W.M. Yen in *Proceedings of VII<sup>th</sup> Phonon Scattering in Condensed Matter VII*, M.Meismu and R.D. Polil, eds., Springer Series in Solid State Studies, vol. 112 (Springer Verlag, 1993) pp.397-398.

"The Performance of a Cr<sup>4+</sup>:YAG Laser in the NIR," W. Jia, H. Eilers, W.M. Dennis, W.M. Yen and A. Shestakov, *Proceedings of Advanced Solid State Lasers Topical Meeting*, L.L. Claire and A. Pinto, eds., OSA Proceedings, vol. 13, pp. 31-33 (1992).

"On the Optical Center in Cr<sup>4+</sup> Doped YAG," H. Eilers, U. Hömmerich, S.M. Jacobsen and W.M. Yen, *Proceedings of the Advanced Solid State Laser*, A.A. Pinto and T.Y. Fau, eds., OSA Proceedings 15, pp.437-440 (1993), (OAS, Washington, D.C.).

"Spectroscopic Investigations of the NIR Center in Cr Doped Y<sub>2</sub>SiO<sub>5</sub>," U. Hömmerich, H. Eilers, W.M. Yen, W. Jia and Y. Wang, *ibid*. 15, pp.444-445 (1993).

"Computer Simulations of Optical Dephasing of Paramagnetic Ions," R.S. Meltzer and D.P. Landau, in *Computer Simulations Studies in Condensed Matter Physics V*, (eds. D.P. Landau, K.K. Mon and H.-B. Schüttler, Springer-Verlag, Berlin, 1993).

#### Non-Refereed Papers

"Yellow and Green Upconversion in Nd:YLF<sub>4</sub>," A.M. Novo-Gradac, S.M. Jacobsen and W.M. Yen, Bull. Am. Phys. Soc. <u>37</u>, 1188 (1992) paper A14 (OAS paper MJ4).

"Laser Spectroscopy Studies of Cr<sup>4+</sup> Centers," S.M. Jacobsen, H. Eilers, K.R. Hoffman and W.M. Yen, *ibid.* 37, 1193 (1992) paper B1-1 (OAS paper MJJ1) (invited).

"Spectroscopy of the Cr,Ca:YAG NIR Laser Center," K.R. Hoffman, U. Hömmerich, S.M. Jacobsen and W.M. Yen, Program of the 182<sup>nd</sup> ECS Annual Meeting, Toronto, Ontario, Oct. 11-16, 1992, pp. 447C, paper 588.

"Optical Saturation Effects in Cr,Ca:YAG," H. Eilers, K.R. Hoffman, W.M. Dennis, S.M. Jacobsen and W.M. Yen, *ibid*. 37, pp. 450C, paper 614.

"Energy Transfer Upconversion Processes in YLiF<sub>4</sub>:Nd<sup>3+</sup>," Bull. Am. Phys. Soc. <u>38</u>, (1994).

"Cross Relaxation Energy Transfer in Tm<sup>3+</sup>:Y<sub>2</sub>O<sub>3</sub>," J.M. Dyson, W.M. Dennis and W.M. Yen, *ibid.* 38, (1994).